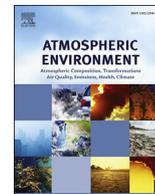




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## Source contributions of urban PM<sub>2.5</sub> in the Beijing–Tianjin–Hebei region: Changes between 2006 and 2013 and relative impacts of emissions and meteorology



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### HIGHLIGHTS

- PM<sub>2.5</sub> in Northern China is apportioned to different categories and regions.
- Dominant roles of industry and residential for fine particles are identified.
- Regional inflow plays a significant role in Tianjin, and central and southern Hebei.
- Meteorology may be as important as emission to change of source contributions.

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### ABSTRACT

Anthropogenic emissions in China have been controlled for years to improve ambient air quality. However, severe haze events caused by atmospheric aerosols with aerodynamic diameter less than or equal to 2.5 μm (PM<sub>2.5</sub>) have continued to occur, especially in the Beijing–Tianjin–Hebei (BTH) region. The Chinese government has set an ambitious goal to reduce urban PM<sub>2.5</sub> concentrations by 25% in BTH by 2017 relative to the 2012 levels. Source apportionment (SA) is necessary to the development of the effective emission control strategies. In this work, the Comprehensive Air Quality Model with extensions (CAMx) with the Particulate Source Apportionment Technology (PSAT) is applied to the China domain for the years 2006 and 2013. Ambient surface concentrations of PM<sub>2.5</sub> and its components are generally well reproduced. To quantify the contributions of each emission category or region to PM<sub>2.5</sub> in BTH, the total emissions are divided into 7 emission categories and 11 source regions. The source contributions determined in this work are generally consistent with results from previous work. In 2013, the industrial (44%) and residential (27%) sectors are the dominant contributors to urban PM<sub>2.5</sub> in BTH. The residential sector is the largest contributor in winter; the industry sector dominates in other seasons. A slight increasing trend (+3% for industry and +6% for residential) is found in 2013 relative to 2006, necessitating more attention to these two sectors. Local emissions make the largest contribution (40%–60%) for all receptors. Change of source contribution of PM<sub>2.5</sub> in Beijing and northern Hebei are dominated by change of local emission. However, for Tianjin, and central and southern Hebei, change of meteorology condition are as important as change of emission, because regional inflow in these areas is more important than in Beijing and northern Hebei and can increase under unfavorable weather conditions, indicating a strong need for regional joint emission control efforts. The results in this study enhance the quantitative understanding of the source–receptor relationships and provide an important basis for policymaking to advance the control of PM<sub>2.5</sub> pollution in China.

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## 1. Introduction

High concentrations of fine particulate matter (defined as particles with aerodynamic diameter less than or equal to 2.5  $\mu\text{m}$ , or  $\text{PM}_{2.5}$ ) is the most severe atmospheric pollution problem in many regions of China, with the Beijing–Tianjin–Hebei (BTH) region the most polluted area. During the haze events of January 2013, eight out of the 10 most polluted cities in the country were in BTH, including Xingtai, Shijiazhuang, Baoding, Handan, Langfang, Hengshui, Tangshan, and Beijing (Wang et al., 2014a). These cities were among the worst-ranked cities by seasonal or annual air quality index during 2013 and 2014. Effective emissions control requires the understanding of  $\text{PM}_{2.5}$  sources and relative contributions. However,  $\text{PM}_{2.5}$  consists of very complex chemical components, which may be generated via several paths. Primary particles are emitted directly from anthropogenic (e.g., industry, power plants, and vehicles) or natural (e.g., outdoor biomass burning and dust storms) sources. Secondary particles are produced in the atmosphere from precursor gases (e.g., sulfur dioxide ( $\text{SO}_2$ ), nitrogen oxides ( $\text{NO}_x$ ), ammonia ( $\text{NH}_3$ ), and volatile organic components (VOCs)) emitted from sources similar to those for primary particles.

Quantitative assessment of sources of  $\text{PM}_{2.5}$  and its components and their contributions (also known as source apportionment (SA)) is a prerequisite to the formulation of the effective emission control strategies. Source apportionment of  $\text{PM}_{2.5}$  in support of emission control strategies needs to meet two requirements. First,  $\text{PM}_{2.5}$  must be fully apportioned to sources because total  $\text{PM}_{2.5}$  is the target of control. Second, the locations of sources must be identified to quantify the role of pollution transport.

Various methods have been developed for SA; several of them are widely used. Chemical Mass Balance (CMB; Watson et al., 1984) is a receptor model that apportions observed pollution concentrations to sources with known source profiles. It assumes that the observed concentrations can be adequately explained by linear combination of a number of sources with fixed source profiles and variable source strengths in time. Other receptor-based approaches with the same assumption, e.g., Positive Matrix Factorization (PMF, Paatero and Tapper, 1994) and UNMIX (Hopke, 2003), are used when source profiles are lacking. These approaches solve the long-term observations with statistical methods and require the user to identify source type from the result, using knowledge of source characteristics. Although receptor models have substantial efficiency in calculation and good performance in SA of primary species, they fail to apportion secondary particles or quantify the contribution of regional transport (Burr and Zhang, 2011a; Lin and Wen, 2015). Trajectory-based methods, e.g., the Potential Source Contribution Function (PSCF; Cheng et al., 1993; Zhang et al., 2013) and Effective Emission Intensity (EEI; Lu et al., 2012), combine gridded emission and back trajectories to estimate regional source contributions. Again, trajectory-based methods are not suitable for SA of secondary particles because they only treat simple chemistry.

The chemical transport model (CTM) has the potential to deal with regional SA and SA of secondary particles, because both the transport and formation of secondary particles are considered. The simplest method is to turn off emission sources of interest one-by-one in each simulation and estimate their contributions from differences between results of these sensitivity simulations and those of the base one. Such a method, the so-called Brute Force Method (BFM) or zero-out method, has been widely used in source sensitivity studies of major pollutants worldwide (e.g., Burr and Zhang, 2011a, b and Wang et al., 2014a).

Source-oriented methods have been developed to obtain source contributions with a single simulation, by tagging species of interest with their sources through all processes in the host model. The source-oriented external mixture (SOEM; Ying and Kleeman, 2006) method is an example. Although SOEM can accurately perform SA, its calculation efficiency is a challenge because the burden of gas-phase chemistry substantially increases with additional source species. The Particulate Source Apportionment Technology (PSAT) does not simulate additional processes of transport, chemistry or deposition for these source species. Instead, changes of such species in these processes are computed in proportion to the change of the original species. PSAT has a much lower burden of gas-phase chemistry calculation than SOEM, and is, therefore, an efficient source tagging method. The Comprehensive Air Quality Model with extensions (CAMx)/PSAT (Yarwood et al., 2005b; Wagstrom et al., 2008) is maintained by ENVIRON and has been updated by both developers and CAMx users. The capability of CAMx/PSAT has been demonstrated by comparing its results with those of other methods (Burr and Zhang, 2011a, b; Bove et al., 2014) and through its wide application in different regions of the world (Bedogni et al., 2008; Wagstrom and Pandis, 2011; Wu et al., 2013; Skyllakou et al., 2014). Several studies have compared results of PSAT with other SA methods. Burr and Zhang (2011a, b) compared different category source contributions between PSAT and BFM. They reported that the two approaches agreed well for primary species but produced significantly different results for secondary species owing to the consideration of nonlinear effects in the BFM but not in PSAT. Bove et al. (2014) compared SA results from PSAT and a receptor-based method, PMF, by reconstructing emission sectors in the PSAT result. Although results from the two methods agreed well for some categories, large uncertainties existed in this comparison.

Several studies have focused on the source contribution of  $\text{PM}_{2.5}$  in China. Wang et al. (2014b) used CAMx-PSAT in SA research in Shanghai, focusing on source contributions in five emissions categories and from local and surrounding areas. Wang et al. (2014a) applied the BFM to a Chinese domain to study source contributions in three cities in the Hebei Province, concluding that local emissions from southern Hebei contributed to > 60% of  $\text{PM}_{2.5}$  in January 2013 for all three cities in the area.

While previous studies provided valuable knowledge of SA of PM, the source contributions will change with changing emissions and may also be affected by a changing climate. Zhang et al. (2014) found large differences of source contributions to  $\text{PM}_{2.5}$  in the United States (U.S.) between 1999 and 2005, consistent with changes in emissions and/or their relative importance between the two years. Anthropogenic emissions in China have changed rapidly since the 1990s relative to those in developed countries (such as the U.S.) because of increasing population, urbanization, and emission controls. To provide timely information for emission control strategies, SA of  $\text{PM}_{2.5}$  of China must be updated as frequently as the changes in emissions to quantify the up-to-date source–receptor relationships and should be performed periodically in conjunction with the major updates in emission inventories.

The goal of this work is to evaluate the change in SA of  $\text{PM}_{2.5}$  in the BTH region from 2006 to 2013 to support the adjustment of emission control policies for the region based on new features of source contributions. To achieve this goal, a regional CTM with the PSAT probing tool is applied over the China domain for two full years: 2006 and 2013. In Section 2, the methods used are described. The simulated results are evaluated in Section 3. SA results for 2006 and 2013 are presented and compared in Section 4. Finally, summarizes the main findings are summarized in Section 5.

## 2. Methodology

### 2.1. Model description and configuration

CAMx is an offline 3-D CTM (ENVIRON, 2013). CAMx v.6.0 is used in this work. In CAMx 6.0, the 2005 Carbon Bond (CB05; Yarwood et al., 2005a) mechanism is used for gas-phase chemistry. The aqueous-phase scheme in the Regional Acid Deposition Model (RADM-AQ) (Chang et al., 1987) scheme is used for aqueous-phase oxidation. Atmospheric aerosols, including sulfate ( $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ), ammonium ( $\text{NH}_4^+$ ), elemental carbon (EC), primary organic carbon (POC), secondary organic carbon (SOC), other unknown particulate matter (other), and mineral dust are simulated using a static two-mode (fine and coarse modes) scheme in CAMx. In this scheme, the fine mode includes  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , EC, POC, and SOC, and both modes include “other” and dust. ISORROPIA (Nenes et al., 1999) is used to simulate inorganic aerosol thermodynamic equilibrium. Dry deposition of aerosols and gases are calculated following Zhang et al. (2001, 2003), respectively. Wet deposition of aerosols and gases are calculated following Seinfeld and Pandis (1998). A detailed description of CAMx can be found in the CAMx 6.0 User's guide (ENVIRON, 2013).

As a species tagging method, PSAT labels emission categories and source regions for particles of interest designed by the user. It then tracks these source species in model integration. After each chemical or physical process (e.g., chemical reaction, deposition, and transport), these species are updated by apportioning the change of corresponding species in the host model (CAMx) to each source. As a result, source information of each selected species on each grid at each time step are delivered and evolved, during which the simulation of the host model is unaffected.

The horizontal resolution is 36-km. The model domain stretches from 68 to 152°E and 10–54°N, covering the whole mainland of China, and the vertical resolution is 23 layers from the surface up to 100 hPa with 10 layers below 3-km. The meteorological fields simulated by the Weather Research and Forecast (WRF) model version 3.5.1 are used to drive CAMx. WRF simulations use the same domain projection, and horizontal and vertical resolutions as those used for CAMx. The domain size of the WRF simulation is designed to be three grid cells larger than that of CAMx on each boundary to ensure a reasonable meteorological boundary condition. The configuration of the WRF simulations is described in Zheng et al. (2015). Initial and boundary conditions (ICBC) of chemical species are interpolated from the output of GEOS-Chem model (Bey et al., 2001; Geng et al., 2015). One-week spin-up is used to minimize the influence of initial conditions on model results. The anthropogenic emissions are based on the Multi-resolution Emission Inventory for China (MEIC, <http://www.meicmodel.org>). The major source categories for these emissions are further described in Section 2.2. Outdoor biomass burning emissions includes forest and grassland fires and crop burning in fields (Huang et al., 2012). The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther et al., 2012) is used to provide biogenic emissions. The dust emissions are generated offline using the dust emission module of Zender et al. (2003) modified by Wang et al. (2012) as implemented in an online-coupled meteorology-chemistry model, WRF with chemistry (WRF/Chem) with physics packages from the Community Atmospheric Model version 5 (WRF-CAM5) (Zhang et al., 2015).

### 2.2. Simulation design

The selected six emission categories include five anthropogenic sectors of the MEIC; industry, power plant, residential, transportation, and agriculture, and natural sources. Natural sources include outdoor biomass burning, biogenic emissions, and mineral

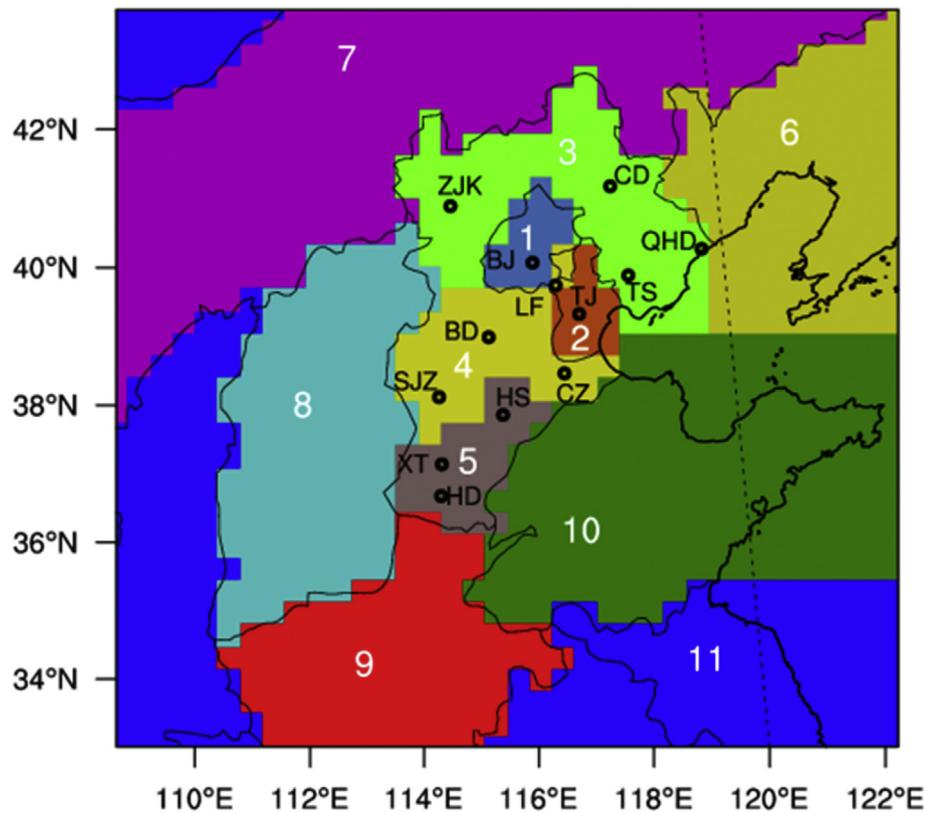
dust. To investigate the role of regional transport, the entire domain is separated into 11 source regions (Fig. 1); Beijing, Tianjin, Northern Hebei (N. Hebei), Central Hebei (C. Hebei), Southern Hebei (S. Hebei), Liaoning, Inner Mongolia, Shanxi, Henan, Shandong, and other areas. Including initial and boundary conditions, there are 68 emission source groups considered in the model ( $[6 \text{ sources} \times 11 \text{ source regions}] + 2 = 68$ ). Thirteen cities in the BTH region are defined as receptors, including Beijing (BJ), Tianjin (TJ), Tangshan (TS), Qinhuangdao (QHD), Chengde (CD), Zhangjiakou (ZJK), Shijiazhuang (SJZ), Baoding (BD), Langfang (LF), Cangzhou (CZ), Hengshui (HS), Xingtai (XT) and Handan (HD). In this work, we use the grid cell at the center of each city to represent the receptor (There are alternative ways to define receptors, such as receptor area. Impact of receptor definition is discussed in Section 4 of Supplemental Materials). Regional SA of urban  $\text{PM}_{2.5}$  is defined as the average SA of  $\text{PM}_{2.5}$  at the centers of all cities in each source region. For instance, SA of urban  $\text{PM}_{2.5}$  for the S. Hebei region refers to the average SA in the cities of Hengshui, Xingtai, and Handan.

CAMx modeling simulations are conducted for 2006 and 2013 with corresponding emissions and meteorological conditions (referred to as E06M06 and E13M13, respectively, see Table 1). The difference between E13M13 and E06M06 reflects the combined impact of changes in emissions and meteorological conditions. The third simulations used 2006 emissions and 2013 meteorology (E06M13). Then the differences between E06M13 and E06M06 represent the effect of changes in meteorological conditions between 2006 and 2013, while the differences between E13M13 and E06M13 (E13M13 minus E06M13) represent the effect of emission changes between two years. All three simulations are conducted for 1 month in each season (January, April, July, and October) to examine the seasonality of model predictions and the resulting SA.

## 3. Model evaluation

Several observational datasets are used to evaluate meteorological and chemical prediction in this study. Datasets and protocol used in this evaluation are described in Supporting Materials. Temperature and relative humidity are generally well reproduced, except that July temperature is underpredicted for both years, which may be affected by predictions of cloud fraction. The wind speed is slightly overpredicted, and, for each season, the U and V components of the wind speed from the prevailing wind direction perform much better than those from the opposite directions. Precipitation is generally underpredicted with Normalized Mean Bias (NMB) from -7.8% to -43.5% and Normalized Mean Error (NME) from 97.4% to 114.1%. Detail performance of meteorological prediction is discussed in Supporting Materials.

Table 2 shows the performance of simulated  $\text{PM}_{2.5}$  for both years. Monthly mean surface concentrations of  $\text{PM}_{2.5}$  in 2006 are comparable to observations, with NMB from -13.5% to 20.1% and NME from 40.0% to 51.1%. However, the surface concentrations of most  $\text{PM}_{2.5}$  components are not reproduced as well as total  $\text{PM}_{2.5}$ , except for  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  (Table S3). The overall good performances of  $\text{PM}_{2.5}$  in 2006 likely come from the combination of overprediction of anthropogenic  $\text{PM}_{2.5}$  and underprediction of mineral dust. Simulated  $\text{PM}_{2.5}$  also shows generally good performance for 2013, except that it is overpredicted by 71.3% in July. Actually, all short-lived species show larger positive biases in July than in other three months. This overprediction is likely due to uncertainties in MEIC and may be associated with the underprediction of 2-m temperature by WRF. It is noted that the  $\text{PM}_{2.5}$  overprediction do not affect much SA results in the BTH region in this work, as the overprediction mainly occurs in southern China. Detailed performance of  $\text{PM}_{2.5}$  components in 2006 and non- $\text{PM}_{2.5}$  species in 2013 are discussed in Supporting Materials.



**Fig. 1.** Map of source regions (numbers in white: 1, Beijing; 2, Tianjin; 3, Northern Hebei; 4, Middle Hebei; 5, Southern Hebei; 6, Liaoning; 7, Inner Mongolia; 8, Shanxi; 9, Henan; 10, Shandong; 11, other areas) and receptors (black circles: BJ-Beijing, TJ-Tianjin; TS-Tangshan; QHD-Qinhuangdao; CD-Chengde; ZJK-Zhangjiakou; BD-Baoding; LF-Langfang; CZ-Cangzhou; SJZ-Shijiazhuang; HS-Hengshui; XT-Xingtai; HD-Handan).

**Table 1**  
Simulation design.

Simulation index	Meteorology	Emission	PSAT
E06M06	2006	2006	On
E13M13	2013	2013	On
E06M13	2013	2006	On

## 4. Results and discussions

### 4.1. SA of $PM_{2.5}$ in 2013

#### 4.1.1. Sectoral SA in 2013

Fig. 2a displays the spatial distribution of annual mean  $PM_{2.5}$  concentrations on the North China Plain (NCP) during 2013. The simulated concentrations of  $PM_{2.5}$  in most areas exceed the  $35 \mu\text{g m}^{-3}$  standard for annual mean concentrations set by the

Ministry of Environment Protection of China.  $PM_{2.5}$  concentrations exceed  $100 \mu\text{g m}^{-3}$  in a large area of BTH, in Henan and Shandong, and tend to peak in cities. The  $PM_{2.5}$  concentrations are apportioned into six source sectors in Fig. 2 (b–g). The ranking of the domain average source contributions of  $PM_{2.5}$  sources from the largest to the smallest is industry, residential, power plant, agriculture, transportation, and natural sources (which is a combination of outdoor biomass burning and mineral dust). In addition to the dominance in magnitude, industry-induced  $PM_{2.5}$  also best explains hotspots in the distribution of total  $PM_{2.5}$ . Compared to industry-induced  $PM_{2.5}$ , residential-induced  $PM_{2.5}$  exhibits a more uniform spatial distribution, because residential emissions are more spatially uniform than industrial emissions. Compared with the former two contributors,  $PM_{2.5}$  attributed to power plants, transportation, and agriculture shows much more uniform distributions, although the emissions of power plants and transportation have obvious hotspots in cities. This probably results from the

**Table 2**  
Performance evaluation of simulated  $PM_{2.5}$ .<sup>a</sup>

	Month	Samples	Corr	MB	ME	RSMB	NMB	NME	MFB	MFE
2006	1	17 <sup>b</sup>	0.76	17.2	38.1	54.3	20.1%	44.6%	-0.6%	48.9%
	4	17	0.64	-9.0	26.6	33.7	-13.5%	40.0%	-20.2%	50.1%
	7	18	0.77	7.7	21.2	26.7	14.8%	40.8%	-3.3%	46.7%
	10	17	0.66	11.6	35.6	47.7	16.7%	51.1%	-14.4%	57.7%
2013	1	61,280	0.48	24.8	76.9	120.0	18.1%	56.3%	8.5%	50.4%
	4	91,057	0.46	7	30.1	47.7	12.1%	52.3%	4.8%	49.3%
	7	96,504	0.43	27.7	40.2	65.6	71.3%	101.9%	34.7%	67.4%
	10	103,734	0.5	-3.1	37.8	63.1	-3.9%	48.7%	-3.2%	49.0%

<sup>a</sup> The units of  $PM_{2.5}$  are  $\mu\text{g m}^{-3}$ .

<sup>b</sup> Sample number for 2006 is actually site number.

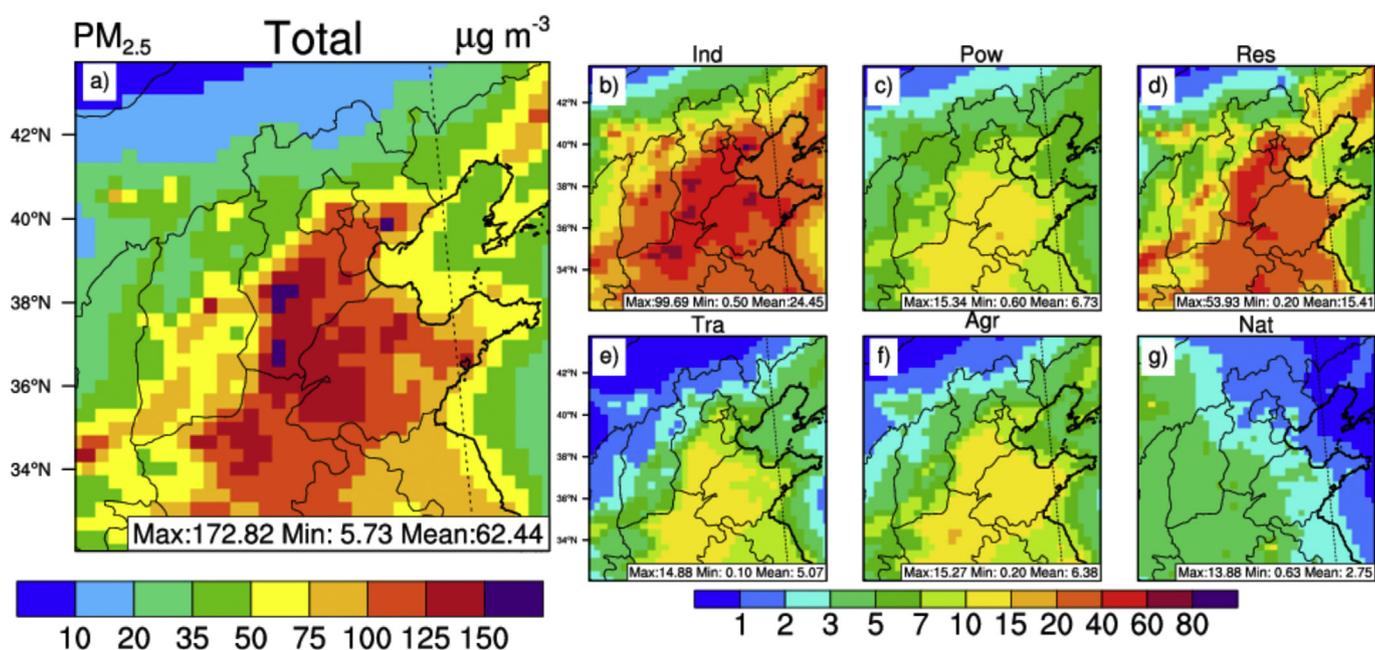


Fig. 2. Simulated spatial distributions of total annual mean PM<sub>2.5</sub> concentrations over the North China Plain in 2013 (a) and contributions from: industry (b), power plants (c), residential (d), transport (e), agriculture (f), and natural sources (g).

combined effect of several factors, including high emission altitudes of power plant sources, uniform distribution of agricultural sources, long lifetime of secondary inorganic particles, and different major emitted species for each sector (e.g., SO<sub>2</sub> and NO<sub>x</sub> for power plants, NO<sub>x</sub> for transportation, and NH<sub>3</sub> for agriculture). Unlike the spatial distributions of SA from anthropogenic sources, for natural PM<sub>2.5</sub>, the impact of dust decreases from west to east, and hotspots are caused by outdoor biomass burning.

Average SA of PM<sub>2.5</sub> in 13 BTH cities is calculated to represent the SA of urban PM<sub>2.5</sub> in the region in 2013 (Fig. 3a–e). The average annual urban PM<sub>2.5</sub> concentration is 117.6 μg m<sup>-3</sup> in BTH. Its contributions are dominated by industry (44%) and residential (30%), followed by power plants (8%), agriculture (8%), and transportation (7%). The remaining 3% are caused by natural sources and boundary transport. Although this ranking is similar to that of the domain average, the relative magnitudes of individual source contributions vary, reflecting the diversity in PM<sub>2.5</sub> spatial distribution for different source categories. The concentration of urban PM<sub>2.5</sub> in BTH peaks in January (221.9 μg m<sup>-3</sup>) with a dominant source contribution from residential (48%), rather than industry (33%), because of substantial residential emissions during winter for heating. The level of urban PM<sub>2.5</sub> during other months is lower and the residential sector contributions (~10%) are similar to those from the power plant, agriculture and transportation sectors. The source contributions of natural sources show some seasonality. Outdoor biomass burning and mineral dust tend to peak in summer and spring, respectively. This tendency is clearer in the SA of PM<sub>10</sub> (Fig. S1) because natural sources are more important than anthropogenic sources for coarse particles.

#### 4.1.2. Regional SA

The above ranking of SA categories provides quantitative information on the sources of total PM<sub>2.5</sub>. However, further understanding of PM<sub>2.5</sub> control, especially at sub regional scale, requires knowledge of PM<sub>2.5</sub> source locations. Fig. 4 shows regional and categorical SA of urban PM<sub>2.5</sub> for five selected receptors. For each receptor, SA labeled as sources within corresponding sub regions is considered as local contributions, and sources with contributions

>2% as non-negligible. As expected, local emissions are the most important contributor for each receptor, accounting for 40%–60% of PM<sub>2.5</sub> (However, when focusing on entire city, e.g. great Beijing, regional contribution could be even higher, as discussed in Section 4 of Supplementary Materials). For most categories, these contributions maximize locally and attenuate with distance. Urban PM<sub>2.5</sub> at all receptors is dominated by either local industry (e.g., in N. Hebei and Tianjin) or local residential contributions (e.g., in Beijing), with important contributions from local agriculture and local transportation.

All five receptors are also influenced by emissions from surrounding areas, across most categories. Regional industry and regional residential sources dominate the emissions inventory of every sub region and hence the inflow contributions. Although local power plant emissions have minor contributions to each receptor, the effect of regional power plants does not attenuate with distance as much as with other sectors and the total contributions of power plants (local plus regional) are as important as those of transportation and agriculture sectors. Anthropogenic source regions typically have a substantial influence on nearby receptors only. One exception is Shandong Province whose influence covers the entire BTH region, because of the large amount of emissions (e.g., industry) in Shandong and the highly efficient transport of PM<sub>2.5</sub> by coastal winds. For natural sources, outdoor biomass burning-induced PM<sub>2.5</sub> are mainly produced locally, whereas fine dust particles originate from the Inner Mongolia and other arid areas.

#### 4.1.3. Comparison with previous results

As shown in Table 3, despite some differences for several sectors, the categorical SA from this work is generally comparable with other work. Those differences are mainly due to different methods employed in different studies. Compared to the results of Wang et al. (2014a) using BFM that accounts for the nonlinear relationship of inorganic aerosols, predicted source contributions to PM<sub>2.5</sub> are larger for power plant and transportation but smaller for agriculture at all three receptors in January 2013. Taking NH<sub>3</sub> as an example, while PSAT accounts for the change in NH<sub>4</sub><sup>+</sup>

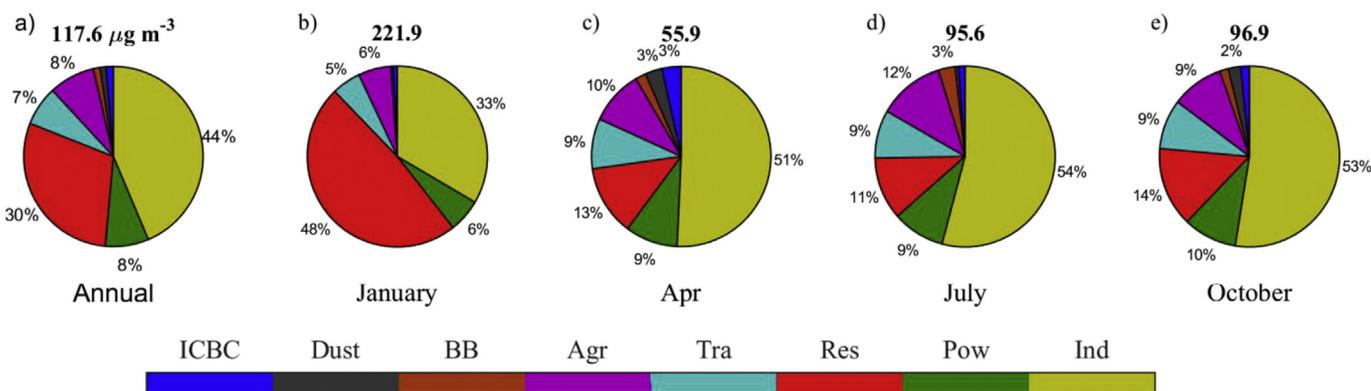


Fig. 3. Source apportionment of urban PM<sub>2.5</sub> of BTH for 2013: annual (a) and seasonal (b–e).

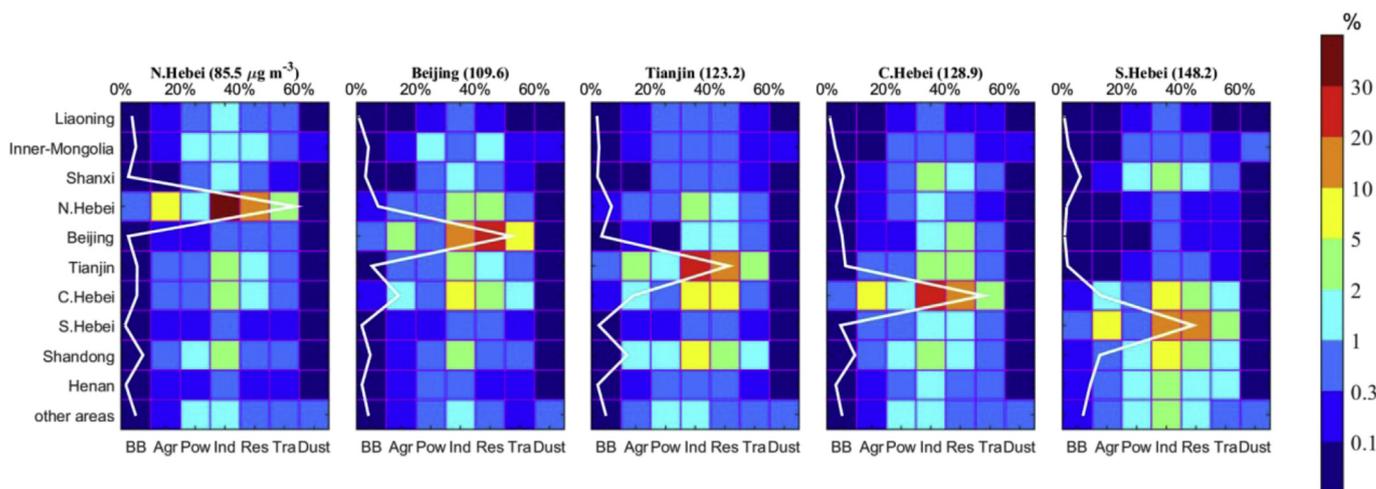


Fig. 4. Annual regional and categorical SA of urban PM<sub>2.5</sub> in Beijing, Tian, N. Hebei, C. Hebei, and S. Hebei for 2013. Colored cells represent percentage contribution of each source region (row) and each source category (column). White lines represent regional contributions (i.e. sum of each row), which is marked by top X-axis. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

concentrations resulting from changes in NH<sub>3</sub> emissions, the BFM also accounts for the related change in SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>, in addition to that for NH<sub>4</sub><sup>+</sup>. What is actually reflected in Wang et al. (2014a) is that inorganic PM<sub>2.5</sub> in winter BTH is more sensitive to the variation of agricultural NH<sub>3</sub> than that of SO<sub>2</sub> or NO<sub>x</sub> emitted mainly from power plants and vehicles.

Mismatching in categorical partitioning is a challenge when comparing SA from receptor-based methods. Therefore, the reconstruction of categories is necessary. The total biomass burning includes residential combined with outdoor biomass burning but neglects residential coal combustion. This is why the predicted biomass burning contributions to EC and POC are generally greater than those reported by Cheng et al. (2013). More complex steps are required to compare with the SA result of Zhang et al. (2013). First, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> from all sources are combined to obtain a factor of secondary inorganic aerosol (SIA). The remaining contribution of transportation is primary particles from vehicles. Following Cheng et al. (2013), 50% of total EC and POC are then considered to represent biomass burning contributions. Mineral dust is renamed as soil dust. Finally, all other contributions are combined into a mixed category, coal combustion plus industrial processes. Comparability between these reconstructed categories and those of Zhang et al. (2013) remains poor. For example, the PMF factor SIA does not actually include all inorganic mass, whereas this is positive in this work, giving greater SIA contribution but smaller

contribution of the mixed category. Meanwhile, owing to a lack of fugitive dust, the simulated soil dust contribution (1%–2%) by this work is much smaller.

Our regional SA results agree well with conclusions from previous studies summarized in Table S5. The predicted city contribution of Shijiazhuang is larger than those of Xingtai and Handan in January 2013, likely due to a higher emission rate and a larger area of Shijiazhuang. These results are consistent with Wang et al. (2014a). The seasonality of local anthropogenic PM<sub>10</sub> in North China shown by Li et al. (2014) with the Nested Air Quality Prediction Model System (NAQPMS) is also well reproduced by the PSAT in this study. Both methods predict extremely small local contributions of PM<sub>10</sub> in the dust season. Among all PM<sub>2.5</sub> species, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> tend to have smaller local contributions. SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> have the smallest local contribution in the warm and cold seasons, respectively, as shown by Ying et al. (2014) and in the present work.

## 4.2. Changes between 2006 and 2013

### 4.2.1. Changes in emissions between years

Anthropogenic emissions in China over the past decade have increased because of increased industrialization and urbanization or decreased because of the adoption of emission controls. Control of primary particles and SO<sub>2</sub>, mainly from power plants, has been

**Table 3**  
Comparison of category contributions with past studies.

Species	Receptor	Source	Period	Previous <sup>a</sup>	This work <sup>b</sup>	Reference & method
PM <sub>2.5</sub>	Shijiazhuang	Industry	January 2013	36.2	37.0	Wang et al., 2014a CMAQ-BFM
		Power plant		0.4	6.0	
		Residential		38.0	45.0	
		Transportation		4.2	5.0	
	Xingtai	Agriculture		14.5	6.0	
		Industry		34.2	30.0	
		Power plant		0.3	6.0	
		Residential		40.5	52.0	
	Handan	Transportation		1.8	5.0	
		Agriculture		16.8	6.0	
		Industry		35.8	30.0	
		Power plant		0.3	6.0	
		Residential		38.1	51.0	
		Transportation		2.9	6.0	
OC	Beijing	Biomass burning	Annual 2011	about 50%	78(77)	Cheng et al., 2013 PMF
EC				about 50%	63(58)	
PM <sub>2.5</sub>	Beijing	Soil dust	Apr 2009–Jan 2010	15.0	1(2)	Zhang et al., 2013 PMF
		Coal Combustion + industry process		43.0	37(38)	
		Biomass burning		12.0	23(20)	
		Primary particles from traffic		4.0	4(5)	
		SIA		26.0	36(35)	

<sup>a</sup> All contributions are in %.

<sup>b</sup> For this study, contributions in 2013 are always listed and, when necessary, corresponding contributions in 2006 are listed in parentheses.

enforced since 2005. NO<sub>x</sub> was not controlled until 2010. Fig. 5 displays the percentage emission changes (from 2006 to 2013) of relevant species in BTH and sub-regions. Focusing on the total amount of emissions in BTH (see black bars in left most panels in Fig. 5), the emissions of SO<sub>2</sub> decrease by 30%, reflecting the effects of emission control on power plants. The emissions of NO<sub>x</sub> increase by 23%, reflecting increased emissions from industry. The emissions of NH<sub>3</sub> decrease by 22%, reflecting decreased emissions from agriculture. Note that there are large uncertainties for this sector. The emissions of EC increase by 10%, reflecting increased emissions from industry partially offset by decreased emissions from transportation. The emissions of POC increase by 7%, largely from an increase of emissions from industry. The emissions of other primary PM<sub>2.5</sub> decrease by 8%, reflecting emission control on power plants.

Although emission changes of all six species in the three sub regions of Hebei are similar to those of the total for BTH, the emission changes in Beijing and Tianjin exhibit very different behaviors. Owing to stricter control measures, the emissions of all species shown in Fig. 5, including NO<sub>x</sub>, EC, and POC, decrease in Beijing. The decline of NO<sub>x</sub> emissions is attributable to decreased emissions from transportation and power plants. Decreases in the emissions of EC and POC are attributable to decreased emission from industry, residential and transportation. Decreased emissions of SO<sub>2</sub> and other primary PM<sub>2.5</sub> in Beijing are of larger magnitudes than those for other sub regions. As shown from the EC and POC panels in Fig. 5, the change in emissions of carbon aerosols in Tianjin is distinct from that in other areas and is driven by changes in the residential sector. The increase of POC in Tianjin is an order of magnitude greater than that of the total in BTH.

#### 4.2.2. Changes of SA from 2006 to 2013

The emission changes described above combined with different meteorological conditions lead to changes in SA in the BTH region. Fig. S2 and Fig. 2 shows the PM<sub>2.5</sub> SA in 2006 and 2013, respectively, and illustrates the dominant role of industry and residential sectors, their increased contributions in most of BTH, and the domain-wide decrease of power-plant related PM<sub>2.5</sub>. Unfortunately, the decrease in power-plant related PM<sub>2.5</sub> is offset by an increase in PM<sub>2.5</sub> from the other sectors, resulting in an overall increase of PM<sub>2.5</sub> over most of the North China Plain. Fig. S3a and Fig. 3a

quantify the changes of contributions to urban PM<sub>2.5</sub> in BTH from industry (from 41% to 44%), residential (24%–30%), and power plants (13%–8%). The remaining panels in Fig. S3 and Fig. 3 show that the seasonal SA changes from 2006 to 2013 generally follow the trends in the annual changes.

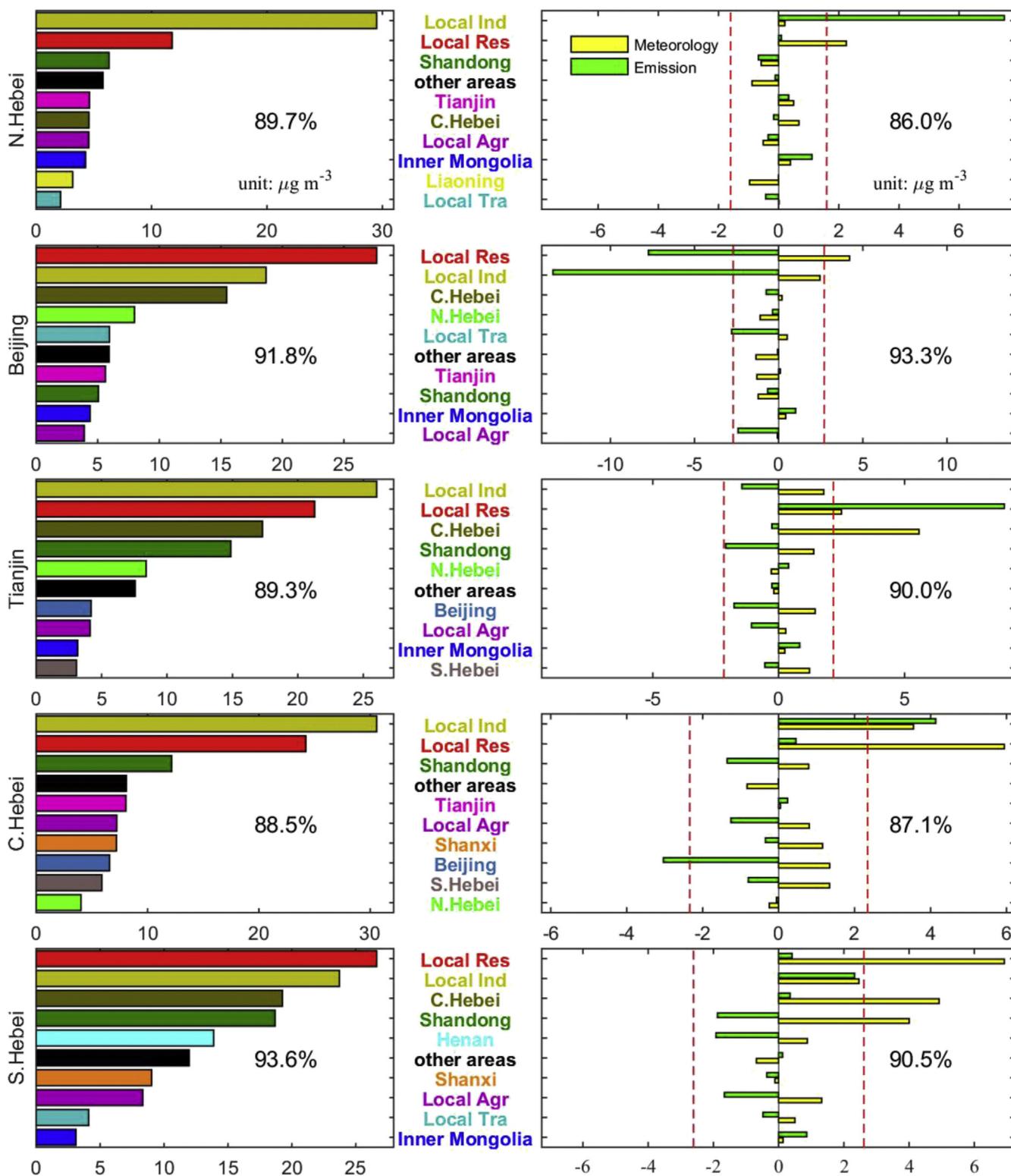
Annual PM<sub>2.5</sub> concentrations in Beijing decreases from 135.3 μg m<sup>-3</sup> in 2006 to 109.6 μg m<sup>-3</sup> in 2013 (Fig. 6a, b), generally consistent with the trend of -3.2 μg m<sup>-3</sup> yr<sup>-1</sup> (-4.3% yr<sup>-1</sup>) during 2005–2013 reported by Zhang et al. (2015). This decrease is a combination of slight decreases in the absolute contributions of all sectors (note that, changes in percentage contribution may not be always consistent with the absolute changes, e.g. residential, the sector with the smallest absolute decrease, shows an increase in its percentage contribution from 31% to 37%). Annual PM<sub>2.5</sub> concentrations in Tianjin, however, increases from 108.7 to 123.2 μg m<sup>-3</sup>. There is a clear decrease from power plants (from 13% in 2006 to 8% in 2013) and increase from residential (20% in 2006 to 31% in 2013), consistent with the absolute contribution from each anthropogenic source (Fig. 6f). In Hebei, the simulated annual PM<sub>2.5</sub> increases from 107.2 to 118.4 μg m<sup>-3</sup>. The power plant contribution decreases from 13% to 8% while the contributions from industry (40%–44%) and residential (23%–29%) increase.

The right panels in Fig. 6 show the PM<sub>2.5</sub> contributions by sector. As discussed above, the simulated industry contribution to PM<sub>2.5</sub> declines in Beijing and rises in Tianjin and Hebei. The decrease in Beijing is attributable to a decrease of other primary PM<sub>2.5</sub> and SO<sub>4</sub><sup>2-</sup>. The increase in Hebei is dominated by POC and SO<sub>4</sub><sup>2-</sup>. Decreased power plant contributions at all three receptors are always dominated by SO<sub>4</sub><sup>2-</sup> (see that for Tianjin in Fig. 6). Increased residential contribution in both Tianjin and Hebei is dominated by POM (Primary Organic Matter, POC multiplied by 1.8) and EC. Decrease of the transportation contribution to PM<sub>2.5</sub> in Beijing is dominated by decreased EC and NO<sub>3</sub><sup>-</sup>. The decline for agriculture in Beijing is caused by decrease of NH<sub>4</sub><sup>+</sup> (figure not shown), NH<sub>4</sub><sup>+</sup> is the only species considered in this sector. Generally, the SA changes discussed above are consistent with emission variations shown in Section 4.2.1, except that there is no evidence for the increase of residential emissions in Hebei, especially POC. This turns out to be an impact of meteorology, which will be discussed subsequently.



To further investigate the changes in major contributors for each receptor, all category and regional contributors for year 2013 are combined into 7 local categories and 10 regions (a total of 17 source groups). The top 10 of the 17 are displayed in Fig. 7 (left) with their

changes due to meteorology and emissions (Fig. 7 right). These top contributors together explain around 90% of total PM<sub>2.5</sub> concentrations for all receptors. Local industry and local residential are always the top two contributors, followed by regional inflow from



**Fig. 7.** (Left) Top 10 contributors of ambient PM<sub>2.5</sub> in 2013 among local categories and regions, and (right) the changes of these contributors from 2006 to 2013 for each receptor. The numbers represent the percentage contribution to either total PM<sub>2.5</sub> concentration (left) or total changes (right) explained by the top 10 contributors together for each receptor. Changes are split into impact of meteorology (yellow) and emission (green) change. Changes that exceed  $\pm 2\%$  of total PM<sub>2.5</sub> in 2006 (dashed lines) are identified. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

surrounding area. Changes of these contributors together explain around 90% of total changes of PM<sub>2.5</sub> concentration between 2006 and 2013.

There are only two notable changes of PM<sub>2.5</sub> in N. Hebei; local industry and residential. The increase in local industry contribution to PM<sub>2.5</sub> is caused by a change in emissions, while the increase in local residential is caused by a change in meteorology. Variations of all regional inflows are negligible (less than 2% of PM<sub>2.5</sub> concentration in E06M06). Similar to N. Hebei, the local residential contribution in Beijing is enhanced by meteorology in 2013. Fortunately, this increase is offset by strict emission controls of sectors in Beijing; the source contributions of local industry, local residential, and local transportation decrease significantly. Again, the role of regional inflow is negligible. As a result, both PM<sub>2.5</sub> concentration and local contribution percentages decline in Beijing from 2006 to 2013.

In contrast to the situation in N. Hebei and Beijing, Tianjin, C. Hebei, and S. Hebei are generally more vulnerable to the impacts of regional inflows. In Tianjin, the source contributions of local residential are enhanced by both emission change and meteorology with the former being more important. In C. Hebei, the local industry contribution is enhanced because of an emission increase, and contributions of both local industry and local residential are enhanced by meteorology. PM<sub>2.5</sub> in C. Hebei also benefits from decreasing emission in Beijing, although this benefit is offset by an increase in other contributors. In S. Hebei, significant changes are increased local residential source contribution and inflows from N. Hebei and Shandong due to meteorological changes. The meteorology in 2013 favors the enhanced contribution of local surface emissions, which is also obvious in comparison of local and non-local contributors (Fig. S8). In all three receptors of Hebei, the contribution of residential emissions to PM<sub>2.5</sub> increases, reflecting changes in meteorological, rather than in emissions (see Fig. 5).

## 5. Conclusions

CAMx/PSATv6.0 has been applied to a Chinese domain to estimate SA of PM<sub>2.5</sub> in the BTH region. Emission categories and source regions are derived in a single simulation. Driven by meteorological fields from the WRF, simulated results capture temporal and spatial characteristics of PM<sub>2.5</sub> and its components across the domain reasonably well. The PM<sub>2.5</sub> concentrations in July 2013 are over-predicted by 70%, which are likely due to uncertainties in MEIC and may be associated with underprediction of 2-m temperature by WRF. This overprediction generally locates in south China and does not notably affect the SA result for BTH. The SA results are generally consistent with conclusions from previous studies.

The industry and residential sectors are found to continue to dominate urban PM<sub>2.5</sub> in BTH from 2006 to 2013; together they contribute to around 70% of urban PM<sub>2.5</sub>. In winter, residential source contributes the most (~50%), followed by industry (~30%). In other seasons, the industry contribution (~50%) is dominant. Contributions to PM<sub>2.5</sub> from industry and residential increase slightly from 2006 to 2013, by 3% and 6%, respectively, reflecting increased emissions of NO<sub>x</sub> and carbon aerosol from the two sectors. When formulating future control policy in BTH, more attention should be given to the residential sector in winter and to industry year round.

Local emission is the most important contributor at all five receptors (40%–60%), although the role of regional inflow is not negligible. Two of the five receptors (Beijing and N. Hebei) have local contribution always >50% for both years. PM<sub>2.5</sub> at these two receptors are generally independent of changes of regional inflow, suggesting that local controls should have a higher priority in improving air quality in Beijing and Northern Hebei than in other receptor areas. PM<sub>2.5</sub> at the other three receptors is more

susceptible to regional inflow. Unfavorable meteorological conditions in 2013 seem to enhance the regional transport of PM<sub>2.5</sub> over the North China Plain, which is demonstrated to be the same important as emission increases from industry and/or the residential sectors. Regional joint actions are clearly an important element of a control strategy.

There are several limitations in this study. In PSAT, a fundamental assumption is that PM is only apportioned to the primary precursor for each PM type, implying that nonlinear relationships among all precursors and secondary PM species are not considered. Therefore, although PSAT gives a true SA result by definition, caution should be executed in using PSAT results to support emission control policy, especially for precursors of secondary inorganic particles. Additionally, source partitioning is currently sector-based, which restricts comparability with results from receptor-based methods and does not provide information on policy of inner-sector emission control. As for all chemical transport modeling work, the uncertainties in meteorology, emission inventories, and formulations and parameterizations used in the 3-D model affect the accuracy of SA results. Although the overall performance of the model in simulating total PM<sub>2.5</sub> is acceptable, the biases in its components may also affect the SA of total PM<sub>2.5</sub> to some extent, because the SA of PM<sub>2.5</sub> components may differ from each other. The contribution of regional inflow is sensitive to the predictions of meteorological variables, e.g., wind speed and boundary layer height. Despite these limitations, the SA results presented in this work provide valuable information regarding the sources and their relative contributions for urban PM<sub>2.5</sub> in the Beijing-Tianjin-Hebei region in 2006 and 2013.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2015.10.048>.

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